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Synthesis of Some New Isolated/Spiro β-Lactam and Thiazolidinone Incorporating Fused Thieno Pyrimidine Derivatives

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Abstract: *Some new spiro β-Lactam and thiazolidinone derivatives were prepared from the reaction of **5** with different aromatic amine to give Schiff bases **6a-c** followed by cycloaddition reaction with chloroacetyl chloride and/or mercaptoacetic acid to give spiro β-Lactam derivatives **7a-c** and spiro thiazolidinone **8a-c**. Also Isolated β-Lactam **11a-c** and Isolated thiazolidinone were prepared by the reaction of Schiff bases **10a-c** with chloroacetyl chloride and/or mercaptoacetic acid, pyrazolo **14,10a-c**. Isoxazolo **16a-c**, pyrimidino **17a c**, pyrimidin thiono **18a-c** Incorporating thieno pyrimidine **9** has been synthesized by cyclocondensation addition reaction of hydrazine hydrate, phenyl hydrazine, hydroxylamine hydrochloride, urea and thiourea*

***Key words**: Spiro β-Lactam, spiro thiazolidinone, thienopyrimidine. Schiff base ,Isolated β-Lactam.*

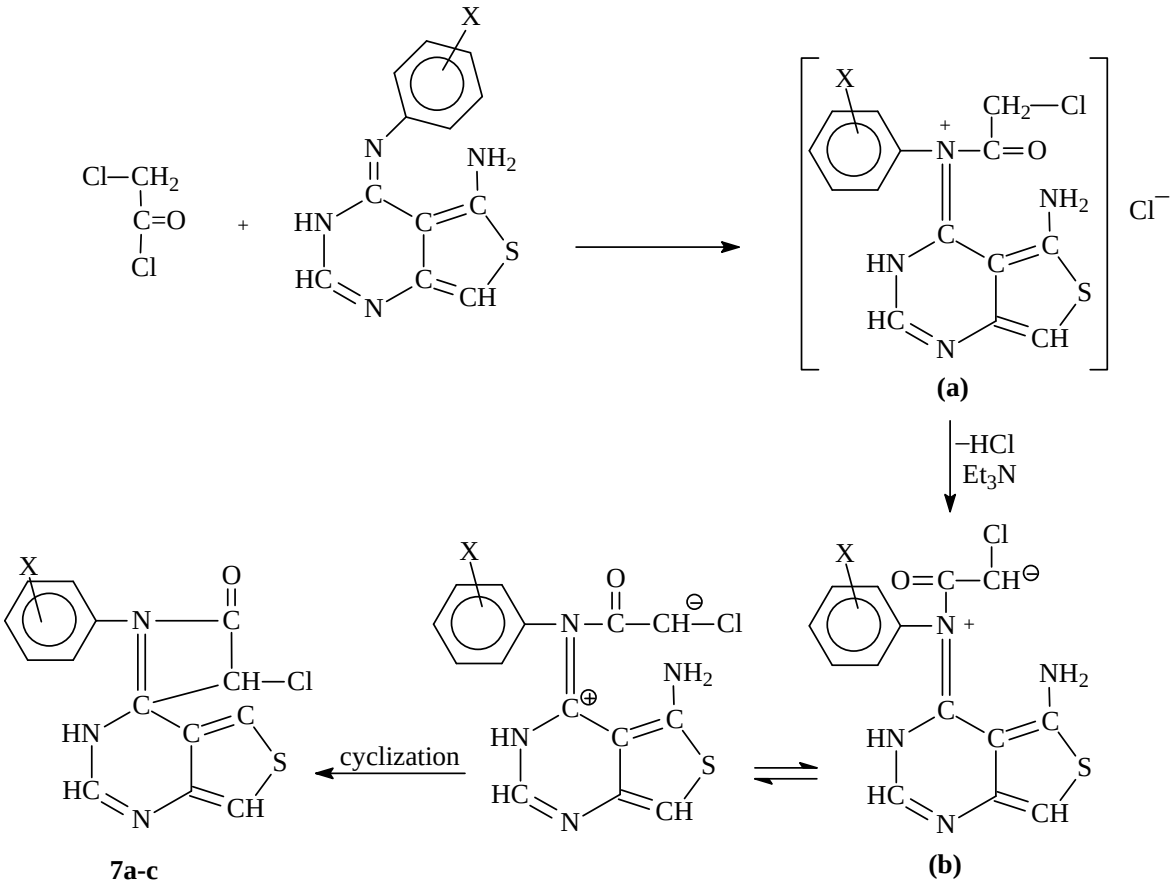
Introduction

The derivatives of fused pyrimidinones have been the focus of great interest over many years due to the fact that many compounds containing a fused pyrimidinone ring play an important role in the biochemistry of the living CeU[1-4]. Pyrazolo[3,4-d]pyrimidine-4-one derivatives also have extremedly rish biological activities because of their structural similarity with purines[5,6], they exhibit excellent antibacterial, antiphlogistic, and antitumor activities[7-10], and they are employed in the treatment of erectile dysfunction in male animals[11-13]. Also β-Lactam antibiotics (e.g. ampicillin, amoxicillin) are traditionally used for the treatment of common bacterial infections in both humans and food-producing animals. B-Lactam residues in foods can result in the development of new strains of bacteria resistant to these antibiotics and in allevgic reactions[14,15].

Result and Discussion

When a mixture of 2,4 diaminozcyarcothiophene 3[16] and conc. Sulfuric acid was stirred at room temperature, hydrolysis of the cyano group took place to give 2,4 diaminothiophene 3-carboxylicacid amide 4 (Scheme 1). The IR spectra of compound 4 revealed the presence of (C=O) at 1645 cm^{-1}, ^{1}H-NMR spectra revealed the presence of 4.25 (brs, 2H, NH_2), 6.65 (s, 2H, $CONH_2$) which support the structure of compound 4. Reaction of compound 4 with triethyl orthoformate under reflux temperature give the corresponding compound 5. The structure of compound 5 was estahlished by IR spectra which revealed the presence of (C=O) at 1700 cm^{-1}, ^{1}H-NMR spectra revealed the presence of NH at δ 12.59 which support the structure of compound 5.

It is very important to know that the formation of Schiff bases corresponding to the newly heterocyclic compounds is the cornerstone in the synthesis of the corresponding spiro and Isolated β-Lactams and thiazolidinone compounds. The activity of the carbonyl group in compound5 render it to react with different aromatic smine to give new Schiff bases **6a-c**. The structures of these newly Synthesized Schiff bases **6a-c** were confirmed by their elemental analysis, IR, ^{1}H-NMR, and mass spectra (cf. Tables I, II). The activity of the azamethine center in compound **6a-c** is more available than the activity of the NH group toward the addition process of chloroacetyl chloride, and this mentioned phenomena is due to the presence of π electron, which makes the foundation of the δ positive and δ negative charge on the carbon and nitrogen atom, respectively, more easy than the presence of this phenomena on the NH group in which the bonding between nitrogen and hydrogen wheather strong according to the nature of this bonding which leads to decreasing of the mobility desire of the hydrogen atom of this pH group[17]. Thus compound **6a-c** reacted with chloroacetyl chloride or mercaptoacetic acid to give spiro β-Lactam and spiro thiazolidinone compound[17] **7a-c** and **8a-c**. The structures of these spiro compounds **7a-c** and **8a-c** were confirmed by their elemental analysis, IR, ^{1}H-NMR and mass spectra (cf. Tables I, II). The formation of spiro azitidine derivatives **7a-c** was suggested to proceed according to the following mechanisms.



Acetylation of thienopyrimidine **5** with one mole equivalent of acetic anhydride yielded the corresponding compound 9. The structures of compound **9** was confirmed by their elemental analysis, IR, ^{1}H-NMR and mass spectra (c.f. Tables I, II).

Compound **9** react with different aromatic nitro so compound to give new Schiff bases **10a-c**. The structures, of **10a-c** were confirmed by their elemental analysis, IR, ^{1}H-NMR and mass spectra (cf. Tables I, II). The activity of azamethine center in compound **10a-c** render it available to react with chloroacetyl chloride and/or mercaptoacetic acid to give new Isolated β-Lactams and thiazolidinone compounds **11a-c** and **12a-c**. The structures of compounds **11a-c** and **12a-c** were confirmed by their elemental analysis, IR, ^{1}H-NMR and mass spectra (cf. Tables I, II). The active methyl group in the new compound **9** condensed with different aromatic aldehydes in a mixture of ethanol and DMF under piperidine as a catalyst to yield the corresponding arilidino **13a-c**. The arylidino derivatives **13a-c**, when interacted with hydrazine hydrate and/or phenyl hydrazine in the presence of acetic acid and/or in ethanol as solvents under piperidine as catalyst, respectively, gave the required N-acetylpyrazolo **14a-c**, and/or N-phenylpyrazolo derivatives **15a-c**, respectively (Scheme 3). The arylidino derivatives **13a-c** when interacted with hydroxylamine hydrochloride in a mixture of ethanol and DMF as solvent under the effect of sodium hydroxide as catalyst,gave the required isooxazolino derivatives **16a-c**. The arylidino derivatives **13a-c**, when interacted with urea and/or thiourea in a mixtgure of ethanol and DMF as solvent under the effect of sodium hydroxide as catalyst, gave the required pyrimidino derivatives **17a-c** or, under the effect of hydrochloric acid as catalyst, gave the required pyrimidinethion derivatives **18a-c**.

Expermental

Melting points are uncorrected, IR spectra were measured as KBr pellets on a pye-unicam sp 1,000 spectrophotometer. ^{1}H-NMR spectra were recorded in ($^2H_6$) dimethylsulfoxide at 200 MHz on a varian Gemini NMR spectrometer using MeSi as an internal reference. Mass spectra were obtained on a Shimadzu GCMS-QP 1000 EX mass spectrometer at 70 eV. Elemental analysis were carried out at the Microanallytical center of Cairo University.

Synthesis of 2,4-diaminothiophene 3-carboxylic acid amide 4:

Compound **3** (0.139 g, 1 mmol) was added portion wise to 20 mL conc. Sulfuric acid, with stirring for 3 h. The reaction mixture was poured into ice-water, neutralized, filtered, dried, and recrystallized from ethanol to give compound **4**.

Synthesis of compound 5:

Compound 4 (0.157 g, 1 mmol) was heated under reflux temperature in 30 mL formic acid for 8 h. The reaction mixture was cooled, poured into water, filtred, dried, and the residue was recrystallized from methanol to give compound **5** (c.f. Tables I, II).

Synthesis of new Schiff bases 6a-c:

Compound 5 (0.1678, 1 mmol) and different aromatic amine (1 mmol) in equindar ratios were dissolved in ethanol, and a few drops of piperidine as catalyst were added; the mixture was refluxed about 10 h. The reaction mixture was allowed to cool at room temperature then filter, washed several times with water, dried and collected, and crystallized from the proper solvent to give **6a-c** (c.f. Tables I, II).

Synthesis of new spiro β-Lactam 7a-c:

A solution of **6a-c** (1 mmol) and chloroacetyl chloride (0.112 g, 1 mmol) in dimethyl formamide (20 mL) in the presence of a few drops of triethylamine. The mixture was refluxed for 8-10. The filtrate was evaporated and ice-water was added, the product was separated, filtred, washed several times with water, and crystallized from the proper solvent to give **7a-c** (c.f. Tables I, II).

Synthesis of new piro thiazolidinone derivatives 8a-c:

A solution of **6a-c** (1 mmol) and mercapto acetic acid (0.0928 g 1 mmol) in dimethyl formamide (30 mL) in the presence of a few drops of triethyl amine. The mixture was refluxed for 9-11 h. The filtrate was evaporated and ice. Water was added, the product was separated, filtred, washed several times with water, and crystallized from the proper solvent to give **8a-c** (c.f. Tables I, II).

Synthesis of compound 9:

Compound **5** (0.167 g, 1 mmol) was heated under reflux temperature in 20 mL glacial acetic acid/acetic anhydride (1:1) for 8 h; then the reaction mixture was cooled, poured into water, filtred, dried, and recrystallized from ethanol to give compound **9** (c.f. Tables I, II).

Synthesis of new Schiff bases 10a-c:

Compound **9** (0.209 g, 1 mmol) and nitro so compound (1 mmol) in equimolar ratios were dissolved in ethanol, and a few drops of piperidine as catalyst were added; the mixture was refluxed for 10-12 h. The reaction mixture was allowed to cool at room temperature then filtrated, washed several times with water, dried and collected, and crystallized from the proper solvent to give **10a-c** (c.f. Tables I, II).

Synthesis of new Isolated β-Lactam 11a-c:

A solution of **10a-c** (1 mmol) in DMF (30 mL) was treated with chloroacetyl chloride (0.112 gm 1 mmol), which was added drop by drop and stirred for 1 h in the presence of triethyl amine catalyst. The reaction mixture was heated under reflux for 10-12 h. (monitored by TLC). The solvent was then evaporated under reduced pressure, and the residue was treated with ice water. The solid product was collected by filtration and crystallized from the propersolvent to give **11a-c** (c.f. Tables I, II).

Synthesis of new Isdolated thiazolidinone derivatives 12a-c:

A solution of **10a-c** (1 mmol) in DMF (30 mL) was treated with mercaptoacetic acid (0.092 g, 1 mmol) in the presence of triethylemine catalyst. The reaction mixture was heated under reflux for 10-12 h. (monitored by TLC). The solvent was then evaporated under reduced pressure, and the residue was treated with ice water. The solid product was collected fy filtration and crystallized from the proper solvent to give **12a-c** (c.f. Tables I, II).

Synthesis of new styryle derivatives 13a-c:

To a solution of **9** (0.209 g, 1 mmol) and different aromatic aldehydes (1 mmol) in a mixtgure of ethand and DMF as solvent and two drops of piperidine as catalyst were added. The reaction mixture was

refluxed for 8-10 h. then left to cool and poured on cold water. The solid product so formed was collected by filtration and crystallized from the proper solvent to give **13a-c** (c.f. Tables I, II).

Synthesis of N-acetyl pyrazolino derivatives 14a-c:

To a solution of **13a-c** (1 mmol) in ethanol as solvent, hydrazine hydrate (0.050 g, 1 mmol) was added followed by glacial acetic acid (10 mL) and the reaction mixture was refluxed for (10-12) h. The reaction mixture was concentrated and coold. The residue were triturated with water, precipitates were separated, filtrated, washed several times with water and crystallized from the proper solvent to give **14a-c** (c.f. Tables I, II).

Synthesis of N-phenylpyrazolino derivatives 15a-c:

To a solution of **13a-c** (1 mmol) in ethanol as solvent phenyl hydrazine (0.108 g, 1 mmol) was added in presence of a few drops of piperidine as catalyst, and the reaction mixture was refluxed for (8-10) h. The reaction mixture was concentrated, triturated with cold water; the crystals were separated. It was filtrated, washed several times with water, and crystallized from the proper solvent to give **15a-c** (c.f. Tables I, II).

Synthesis of N-isooxazolino derivatives 16a-c:

Compound **13a-c** (1 mmol) were refluxed with hydroxylamine hydro-chloride (0.069 g, 1 mmol) in the presence of sodium hydroxide as catalyst and ethanol as solvent for (5-6) h. The reaction mixtrure was filtrated from unreacted materials; the filtrate was triturated with cold water, the product were separated, filtrated, washed several times with water, and crystallized from the proper solvent to give **16a-c** (c.f. Tables I, II).

Synthesis of N-pyrimidino derivatives 17a-c:

Compounds **13a-c** (1 mmol) were refluxed with urea (0.06 g, 1 mmol) in presence of HCl as catalyst and ethanol as solvent for (6-8) h. The reaction mixture was filtrated from unreacted materials; the filtrate was concentrated and triturated with water, the products were separated, filtrated, washed several times with water and crystallized from the proper solvent to give **17a-c** (c.f. Tables I, II).

Synthesis of N-thiopyrimidino derivatives 18a-c:

Compounds **13a-c** (1 mmol) were refluxed with thiourea (0.076 g, 1 mmol) in the presence of sodium hydroxide as catalyst and ethanol as solvent for (6-8) h. The reaction mixture was filtrated; the filtrate was concentrated, triturated with water. The products were separated, filtrated, washed several times with water and crystallized from the proper solvent to give **18a-c** (c.f. Tables I, II).

Table I

Comp. No.	Solvent of Crystallization	m.p.^{o}C	Yield %	M. Formula (m.wt)	Analytical data found/required %					MS (m/z)
					C	H	N	S	Cl	
4	EtOH	275	75	$C_5H_7ON_3S$ (157.18)	38.20 38.19	4.49 4.47	26.73 26.72	20.40 20.39	- -	157
5	MeOH	290	65	$C_6H_5ON_3S$ (167.18)	43.11 43.09	3.01 3.00	25.13 25.12	19.18 19.16	- -	167
6a	EtOH	>300	60	$C_{12}H_{10}ON_4S$ (242.30)	59.49 59.48	4.16 4.15	23.12 23.10	13.23 13.21	- -	242
6b	EtOH	>300	63	$C_{12}H_9O_2N_3S$ (287.29)	50.17 50.16	3.16 3.14	24.38 24.37	11.16 11.14	- -	287
6c	DMF/EtOH	>300	67	$C_{12}H_{10}ON_4S$	55.80 55.78	3.90 3.89	21.69 21.67	12.41 12.40	- -	258
7a	DMF	>300	61	$C_{14}H_{11}ON_4SCl$ (318.79)	52.75 52.74	3.48 3.46	17.58 17.57	10.06 10.04	11.12 11.11	318
7b	DMF/EtOH	>300	59	$C_{14}H_{10}O_3N_5SCl$	46.22 46.21	2.77 2.76	19.25 19.23	8.81 8.80	9.75 9.73	363
7c	DMF/EtOH	>300	60	$C_{14}H_{12}O_2N_4SCl$ (335.79)	50.08 50.06	3.60 3.58	16.68 16.67	9.55 9.54	10.56 10.54	335
8a	DMF	>300	62	$C_{14}H_{12}ON_4S_2$ (316.64)	53.15 53.09	3.82 3.80	17.78 17.77	20.25 20.23	- -	316
8b	DMF	>300	64	$C_{14}H_{11}O_3N_5S_2$ (361.41)	46.53 46.52	3.07 3.06	19.38 19.36	17.74 17.73	- -	361
8c	DMF	>300	61	$C_{14}H_{12}O_2N_4S_2$ (332.00)	50.59 50.57	3.64 3.63	16.86 16.84	19.29 19.28	- -	332
9	EtOH	265	60	$C_8H_{17}O_2N_3S$ (209.22)	45.93 45.92	3.37 3.35	20.08 20.07	15.32 15.31	- -	209
10a	MeOH	>300	62	$C_{18}H_{12}O_2N_4S$ (348.38)	62.06 62.04	3.47 3.46	16.08 16.06	9.20 9.19	- -	348
10b	MeOH	>300	64	$C_{18}H_{12}O_2N_4S$ (348.38)	62.06 62.05	3.47 3.46	16.08 16.07	9.20 9.18	- -	348
10c	MeOH	>300	65	$C_{16}H_{15}O_2N_5S$ (373.45)	51.46 51.44	4.05 4.04	18.75 18.73	8.58 8.57	- -	373
11a	EtOH	>300	61	$C_{20}H_{13}O_3N_4SCl$ (424.87)	56.54 56.53	3.08 3.07	13.19 13.17	7.55 7.54	8.35 8.33	424
11b	EtOH	>300	63	$C_{20}H_{13}O_3N_4SCl$ (424.87)	56.54 56.52	3.08 3.07	13.19 13.18	7.55 7.53	8.35 8.33	424
11c	EtOH	>300	60	$C_{18}H_{16}O_3N_5SCl$ (417.87)	51.74 51.73	3.86 3.84	16.76 16.75	7.67 7.65	8.49 8.48	417
12a	MeOH	>300	59	$C_{20}H_{14}O_3N_4S_2$ (422.47)	56.86 56.85	3.34 3.32	13.26 13.25	15.18 15.16	- -	422
12b	MeOH	>300	62	$C_{20}H_{14}O_3N_4S_2$ (422.47)	56.86 56.84	3.34 3.33	13.26 13.24	15.18 15.17	- -	422
12c	MeOH	>300	61	$C_{18}H_{17}O_3N_5S_2$ (415.48)	52.04 52.03	4.12 4.10	16.86 16.87	15.43 15.42	- -	415
13a	DMF	>300	62	$C_{15}H_{11}O_2N_5S$ (297.33)	60.59 60.57	3.73 3.72	14.13 14.11	10.78 10.76	- -	297

Table 1 (Continued).

Comp. No.	Solvent of Crystallization	m.p.°C	Yield %	M. Formula (m.wt)	Analytical data found/required %					MS (m/z)
13b	DMF	>300	60	$C_{15}H_{11}O_3N_3S$ (313.33)	57.50 57.49	3.54 3.52	13.41 13.40	10.23 10.21	- -	313
13c	DMF	>300	63	$C_{15}H_{10}O_4N_4S$ (342.33)	52.63 52.61	2.94 2.93	16.37 16.35	9.37 9.36	- -	342
14a	MeOH	>300	60	$C_{17}H_{13}O_2N_5S$ (351.38)	58.12 58.11	3.73 3.72	19.93 19.92	9.12 9.10	- -	351
14b	MeOH	>300	62	$C_{17}H_{13}O_3N_5S$ (367.38)	55.58 55.56	3.57 3.56	19.06 19.04	8.73 8.71	- -	367
14c	MeOH	>300	61	$C_{17}H_{12}O_4N_6S$ (396.38)	51.51 51.50	3.05 3.03	21.20 21.19	8.09 8.07	- -	396
15a	EtOH	>300	59	$C_{21}H_{15}ON_5S$ (385.44)	65.44 65.42	3.92 3.91	18.17 18.15	8.32 8.31	- -	385
15b	EtOH	>300	56	$C_{21}H_{15}O_2N_5S$ (401.44)	62.83 62.82	3.77 3.75	17.45 17.44	7.99 7.97	- -	401
15c	EtOH	>300	55	$C_{21}H_{14}O_3N_6S$ (430.44)	58.60 58.59	3.28 3.27	19.52 19.50	7.45 7.44	- -	430
16a	MeOH	>300	54	$C_{15}H_{10}O_2N_4S$ (310.34)	58.06 58.04	3.25 3.24	18.05 18.03	10.33 10.32	- -	310
16b	MeOH	>300	55	$C_{15}H_{10}O_3N_4S$ (326.33)	55.21 55.20	3.09 3.07	17.17 17.16	9.82 9.80	- -	326
16c	MeOH	>300	52	$C_{15}H_9O_4N_5S$ (355.33)	50.70 50.69	2.55 2.54	19.71 19.69	9.02 9.00	- -	355
17a	EtOH	>300	56	$C_{16}H_{11}O_2N_5S$ (337.35)	56.97 56.96	3.29 3.27	4.15 4.14	9.50 9.48	- -	337
17b	DMF	>300	57	$C_{16}H_{11}O_3N_5S$ (353.35)	54.39 54.38	3.14 3.12	19.82 19.81	9.07 9.05	- -	353
17c	DMF/EtOH	>300	54	$C_{16}H_{10}O_4N_6S$ (382.35)	50.26 50.24	2.62 2.61	21.98 21.96	8.38 8.37	- -	382
18a	EtOH	>300	55	$C_{16}H_{10}O_4N_5S_2$ (353.42)	54.38 54.37	3.14 3.12	19.82 19.81	18.14 18.12	- -	353
18b	EtOH	>300	53	$C_{16}H_{11}O_2N_5S_2$ (369.41)	52.02 52.00	3.00 2.99	18.96 18.94	17.36 17.35	- -	369
18c	EtOH	>300	52	$C_{16}H_{10}O_3N_6S_2$ (398.41)	48.24 48.23	2.53 2.51	21.09 21.08	16.09 16.07	- -	398

Table II

Compound No.	IR υ_{max}/cm^{-1}	^{1}H-NMR (DMSO) ppm
4	1645 (C=O), 3100-3400 (NH_2).	δ 4.25 (brs, 2H, NH_2), δ 6.65 (s, 2H, $CONH_2$), 8.00-7.01 (m, 3H, aromatic protons).
5	1700 (C=O), 3100-3400 (NH, NH_2).	δ 8.01-7.01 (m, 5H, aromatic protons), δ 12.59 (br, 1H, NH).
6a	1585 (C=N), 3100-3450 (NH, NH_2).	δ 8.10-7.01 (m, 9H, aromatic protons), δ 11.5 (brs, NH).
6b	1595 (C=N), 3100-3400 (NH, NH_2).	δ 7.01-7.01 (m, 8H, aromatic protons), δ 12 (brs, NH).
6c	1590 (C=N), 3100-3450 (NH, NH_2, OH).	δ 8.01, 7.01 (m, 9H aromatic protons), δ 10.5 (brs, NH).
7a	1650-1725 (C=O), 3100-3400 (NH, NH_2).	δ 8.01-7.01 (m, 10 Haromatic protons), δ 10.45 (brs, NH).
7b	1655-1725 (C=O), 3100-3400 (NH, NH_2).	δ 8.01-7.01 (m, 9H aromatic protons), δ 10.85 (brs, NH).
7c	1655-1720 (C=O), 3100-3450 (NH, NH_2, OH).	δ 8.01-7.01 (m, 11H aromatic protons), 10.99 (brs, NH).
8a	1650-1717 (C=O), 3100-3400 (NH, NH_2).	δ 2.5 (CH_2 of thiazolidinone), δ 8.01-7.01 (m, 9 Haromatic protons), δ 10.35 (brs, NH).
8b	1660-1720 (C=O), 3100-3400 (NH, NH_2).	δ 2.5 (CH_2 of thiazolidinone), δ 8.01-7.01 (m, 8 Haromatic protons), δ 10.6 (brs, NH).
8c	1655-1725 (C=O), 3100-3450 (NH, NH_2).	δ 2.5 (CH_2 of thiazolidinone), δ 8.01-7.01 (m, 9 Haromatic protons), δ 10.45 (brs, NH).
9	1705 (C=O), 1650 (C=O), 3100-3450 (NH_2).	δ 2.15 (s, 3H, CH_3), δ 8.01-7.01 (m, 4H aromatic protons).
10a	1580 (C=N), 1650-1725 (2C=O), 3100-3450 (NH_2, OH).	δ 6.79 (brs, 2H, NH_2), δ 8.01-7.01 (m, 10H aromatic protons).
10b	1585 (C=N), 1655-1720 (2C=O), 3100-3450 (NH_2, OH).	δ 6.59 (brs, 2H, NH_2), δ 8.1-7.01 (m, 10H aromatic protons).
10c	1590 (C=N), 1650-1715 (2C=O), 3100-3400 (NH_2).	δ 1.21 (s, 6H, $2CH_3$), δ 6.12 (brs, 2H, NH_2), 8.1-7.01 (m, 7H aromatic protons).
11a	1650-1720 (3C=O), 3100-3450 (NH_2, OH).	δ 6.13 (brs, 2H, NH_2), δ 8.1-7.01 (m, 11H aromatic protons).
11b	1655-1715 (3C=O), 3100-3450 (NH_2, OH).	δ 5.99 (brs, 2H, NH2), δ 8.1-7.01 (m, 11H aromatic protons).
11c	1660-1715 (3C=O), 3100-3400 (NH_2).	δ 1.25 (s, 6H, $2CH_3$), δ 6.12 (brs, 2H, NH_2), δ 8.1-7.01 (m, 8H aromatic protons).

Table II (Continued)

Compound No.	IR υ_{max}/cm^{-1}	^{1}H-NMR (DMSO) ppm
12a	1665-1720 (3C=O), 3100-3450 (NH2, OH).	δ 2.53 (s, 2H, CH_2 of thiazolidinone), δ 5.95 (brs, 2H, NH_2), δ 8.1-7.01 (m, 10H aromatic protons).
12b	1660-1725 (3c=O), 3100-3450 (NH_2, OH).	δ 2.55 (s, 2H, CH_2 of thiazolidinone), δ 6.01 (brs, 2H, NH_2), δ 8.1-7.01 (m, 10H, aromatic protons).
12c	1655-1720 (3C=O), 3100-3400 (NH_2).	δ 1.22 (s, 6H, $2CH_3$), δ 2.51 (s, 2H, CH_2 of thiazolidinone), δ 5.98 (brs, 2H, NH_2), δ 8.1-7.01 (m, 7H aromatic protons).
13a	1650-1715 (2C=O), 3100-3400 (NH_2).	δ 6.55 (brs, 2H, NH_2), δ 8.1-7.01 (m, 9H, aromatic protons).
13b	1655-1718 (2C=O), 3100-3450 (NH_2, OH).	δ 6.65 (brs, 2H, NH_2), δ 8.1-7.01 (m, 9H aromatic protons).
13c	1665-1725 (2C=O), 3100-3400 (NH_2).	δ 6.75 (brs, 2H, NH_2), δ 8.1-7.01 (m, 8H, aromatic protons).
14a	1670-1725 (2C=O), 3100-3400 (NH_2).	δ 3.31 (s, 3H, COCH3), δ 6.77 (brs, 2H, NH2), 8.1-7.01 (m, 8H, aromatic protons).
14b	1669-1720 (2C=O), 3100-3450 (NH_2, OH).	δ 3.30 (s, 3H, $COCH_3$), δ 6.74 (brs, 2H, NH_2), 8.1-7.01 (m, 8H aromatic protons).
14c	1675-1725 (2C=O), 3100-3450 (NH_2).	δ 3.35 (s, 3H, $COCH_3$), δ 6.79 (brs, 2H, NH_2), 8.1-7.01 (m, 7H, aromatic protons).
15a	1685 (C=O), 3100-3400 (NH_2).	δ 6.58 (brs, 2H, NH_2), δ 8.1-7.01 (m, 13H, aromatic protons).
15b	1690 (C=O), 3100-3450 (NH_2, OH).	δ 6.62 (brs, 2H, NH_2), 8.1-7.01 (m, 13H, aromatic protons).
15c	1700 (C=O), 3100-3400 (NH_2).	δ 6.68 (brs, 2H, NH_2), 8.1-7.01 (m, 12H, aromatic protons).
16a	1684 (C=O), 3100-3400 (NH_2).	δ 6.54 (brs, 2H, NH_2), 8.1-7.01 (m, 8H, aromatic protons).
16b	1689 (C=O), 3100-3450 (NH_2, OH).	δ 6.56 (brs, 2H, NH_2), 8.1-7.01 (m, 8H, aromatic protons).
16c	1695 (C=O), 3100-3400 (NH_2).	δ 6.60 (brs, 2H, NH_2), 8.1-7.01 (m, 7H, aromatic protons).
17a	1685-1715 (2C=O), 3100-3400 (NH, NH_2).	δ 6.1 (brs, 2H, NH_2), 8.1-7.01 (m, 8H, aromatic protons), δ 11.12 (brs, 1H, NH).
17b	1690-1720 (2C=O), 3100-3450 (NH, NH_2, OH).	δ 6.23 (brs, 2H, NH_2), 8.1-7.01 (m, 8H, aromatic protons), δ 11.2 (brs, 1H, NH).
17c	1695-1725 (2C=O), 3100-3400 (NH, NH_2).	δ 6.45 (brs, 2H, NH_2), 8.1-7.01 (m, 7H, aromatic protons), δ 11.35 (brs, 1H, NH).

Table II (Continued).

Compound No.	IR υ_{max}/cm^{-1}	^{1}H-NMR (DMSO) ppm
18a	1686 (C=O), 3100-3400 (NH, NH_2).	δ 6.33 (brs, 2H, NH_2), 8.1-7.01 (m, 8H, aromatic protons), δ 10.95 (brs, 1H, NH).
18b	1689 (C=O), 3100-3450 (NH,NH_2, OH).	δ 6.35 (brs, 2H, NH_2), 8.1-7.01 (m, 8H, aromatic protons), δ 10.98 (brs, 1H, NH).
18c	1693 (C=O), 3100-3400 (NH, NH_2).	δ 6.55 (brs, 2H, NH_2), 8.1-7.01 (m, 7H, aromatic protons), 11.02 (brs, 1H, NH).

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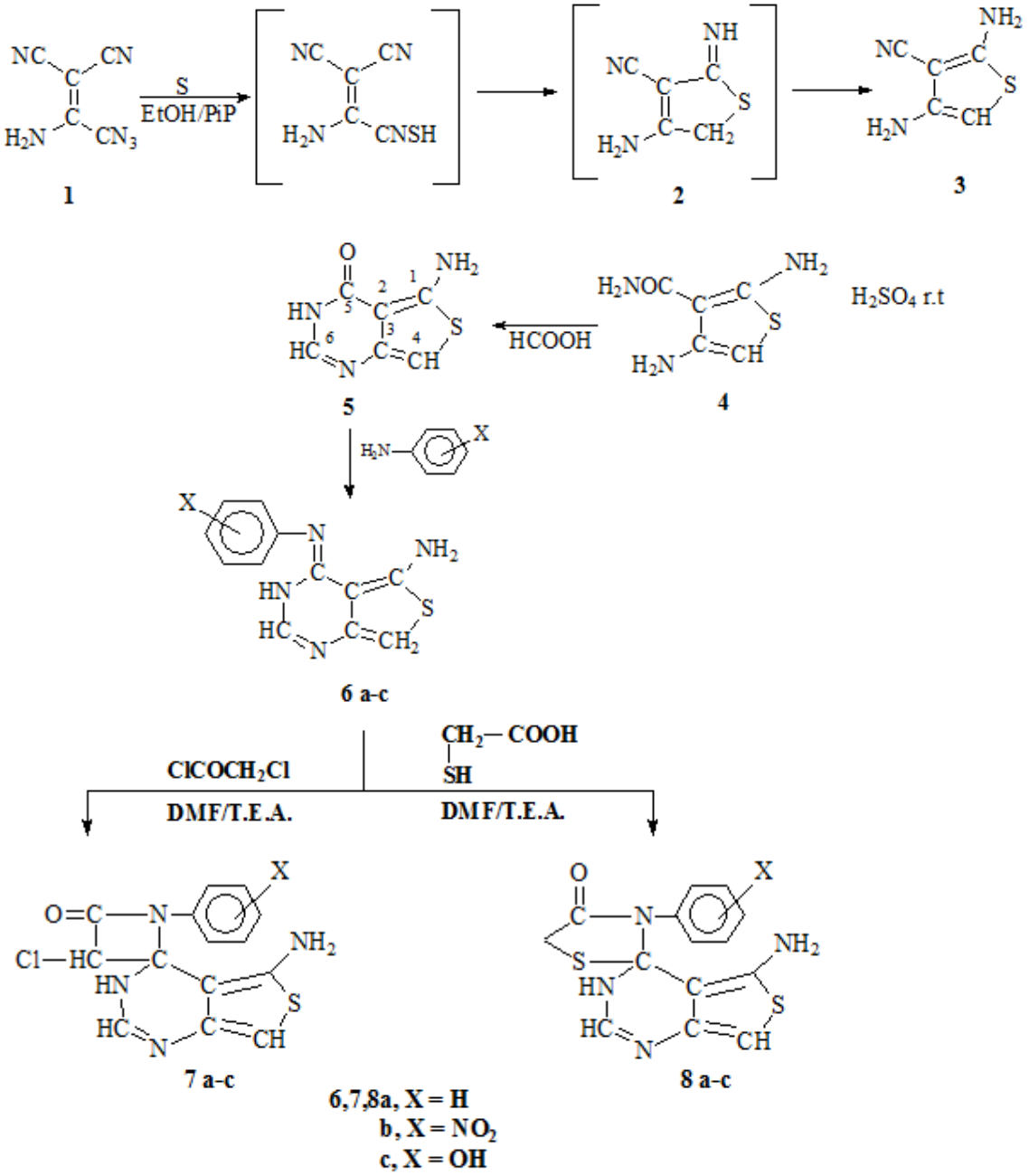
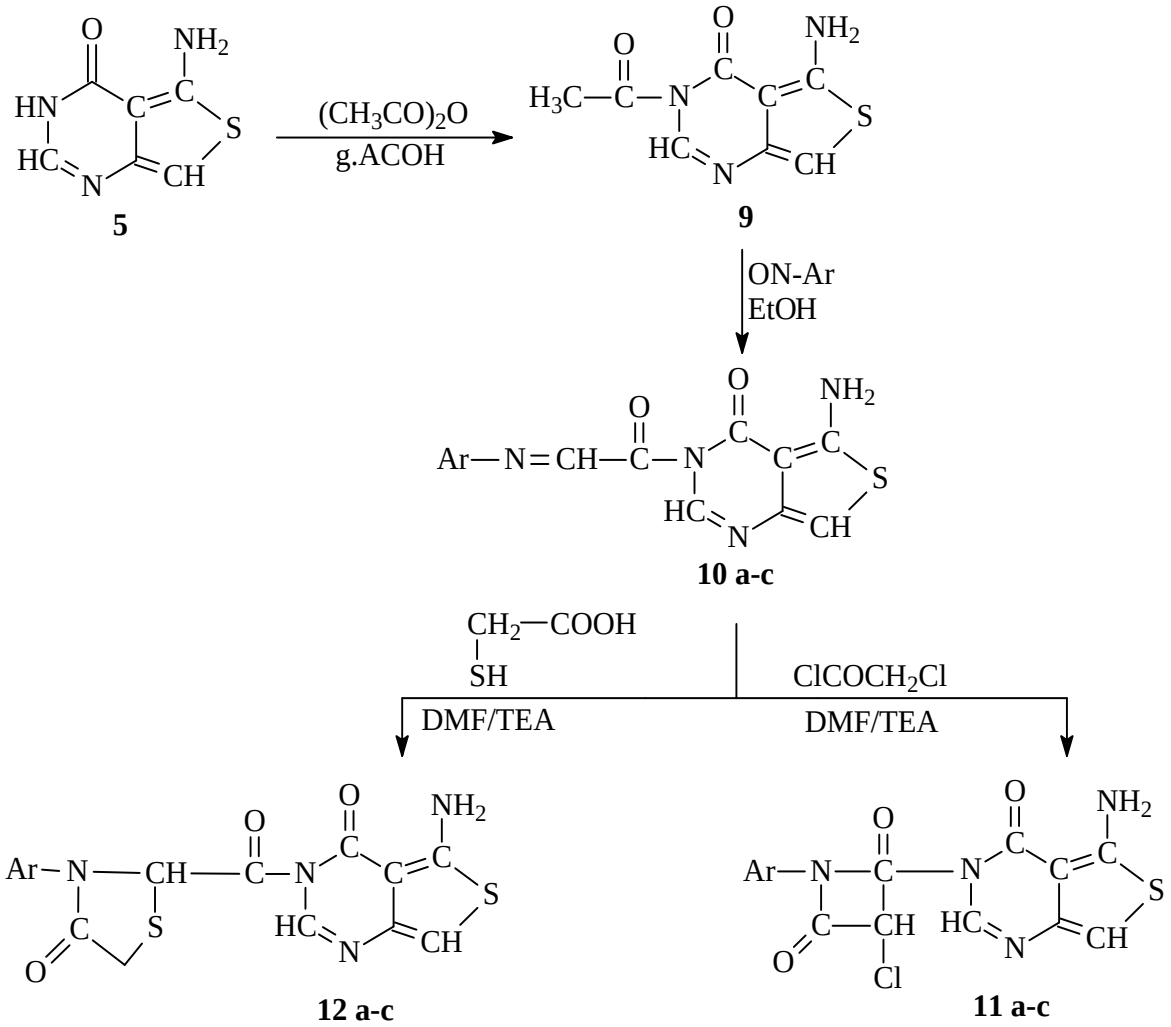
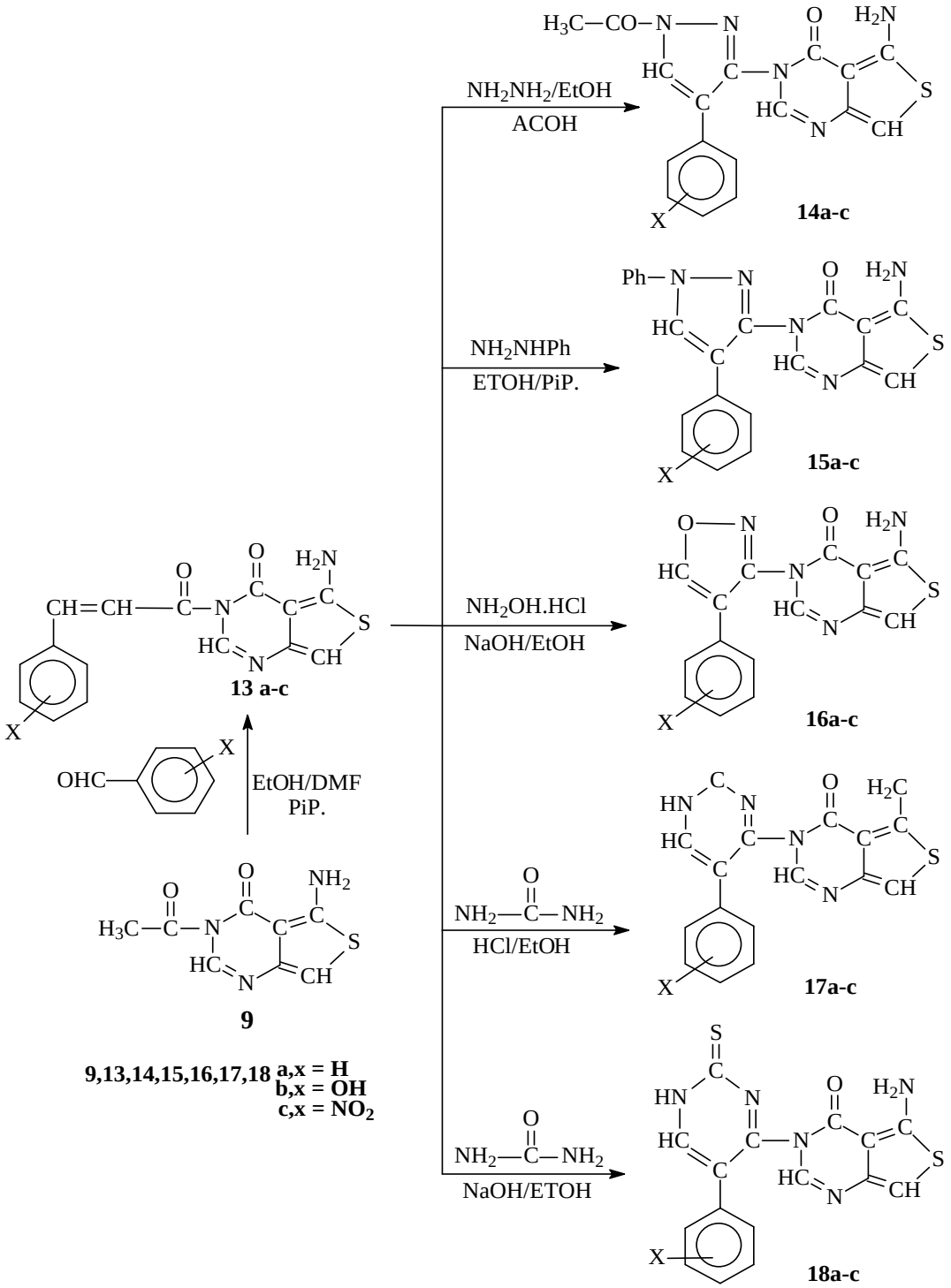

S
EtOH/PiP
HCOOH
H2SO4 r.t
ClCOCH2Cl
DMF/T.E.A.
CH2—COOH
SH
DMF/T.E.A.
1
2
3
4
5
6 a-c
7 a-c
8 a-c
6,7,8a, X = H
b, X = NO2
c, X = OH

Scheme 1



where a, Ar α-nitroso β-naphthol; Ar β-nitroso α-naphthol; c, Ar p-nitroso N,N-dimethylaniline

Scheme2


NH2NH2/EtOH
ACOH
14a-c
NH2NHPh
ETOH/PiP.
15a-c
NH2OH.HCl
NaOH/EtOH
16a-c
NH2—C—NH2
HCl/EtOH
17a-c
NH2—C—NH2
NaOH/ETOH
18a-c
13 a-c
EtOH/DMF
PiP.
9
9,13,14,15,16,17,18 a,x = H
b,x = OH
c,x = NO2

Scheme 3